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14. ABSTRACT
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MATHEMATICAL MODELING, SIMULATION, AND CONTROL OF PHYSICAL PROCESSES

AFOSR F49620-02-1-0132

FINAL TECHNICAL REPORT

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Summary

This research program had as fundamental objectives: to develop a mathematical model of the physical processes attendant to material deposition, particularly pulsed laser deposition, including the ablation, plume dynamics, and film growth, with a focus on eventual automatic control of the process; to develop efficient ways of computing and simulating some of the physical processes of pulsed laser deposition, and to complement experimental results to understand the basic physics and chemistry of the ablation plume; to model the magnetocaloric effect and other magnetoelastic interactions in magnetic materials; to develop computational methods of evaluating mesoscale models of physiological systems (i.e., complex systems); to analyze designs for wing-in-ground-effect transport planes. The goals of this program had an immediate focus on supporting scientists at AFRL/WPAFB, as well as being relevant to long range Air Force *defense-after-next* plans.

The main effort was the development of mathematical models for pulsed laser deposition, including quantifying the relationship between all of the processing variables and

sensors in order to expose the structure of the process as an input-output system. In this effort, considerable attention was given to the physical processes occuring in the ablation plume. For example: How does the plume affect a film deposition? Explain certain observed scaling relationships; i.e., the pressure/distance scaling relationship for optimal grain size. Are there analytic pointers to the cross-over times for the different asymptotic regimes? Concerning the dynamic composition of the plume: how are chemical reactions occuring in the plume? How important is oxide production in the plume vis a vis kinetics on the substrate, and does it depend on process variables? How does the substrate temperature control influence the plume? How does nancluster formation depend on the dynamics of the plume? Can one identify instabilities which play a role in the material fabrication? What practical role does turbulence play in PLD dynamics? Which process outputs can be used for feedback control? Does UV emission give an unambiguous observation of the plume?

Energy flow in the laser/material interaction results in a short-lived plasma and a supersonic plume of material that deposits on a substrate. A model was developed that considered possible ablation mechansisms, both thermal and far-from equilibrium ablation via electronic or photoelectronic avenues. A full mathematical model has allowed us to simulate the ablation plume. Numerical simulations are an important tool in order to help match simple plume models with the spectroscopic observations, which in many cases are the only real-time outputs of the laser ablation process. As a complement to the numerical modeling, we developed analytic models to represent the plume at different scales. These analytic models help explain features found in both experimental and numerical investigations of the plume. These ablation problems are related to hypersonic ablation, particularly the recondensation in the Knudsen layer, and also have important applications to polymer materials, as well as ablative laser propulsion. Nanocluster formation through PLD is a passible path to materials with novel properties. (Condensation occurs in the decaying plume). Whether major cluster formation occurs in the plume or on the substrate or on the substrate is a current point of contention. In this project, we investigated how cluster formation depends on processing parameters.

The magnetocaloric effect in magnetic and superconducting materials was computed in the linearized limit using generalized magnon-phonons. An on-going project is the fabrication of nano-structured magnetic materials in the Wright State laboratory. These superparamagnetic materials, fabricated by electro-deposition using a membrane may also be of use in the future for storage/recording media. This is an unusual real world example where the Heisenberg models of magnetic moments have some validity. We are presently modifying an Ising model and are planning Monte Carlo calculations of the macroscopic moment and the the relaxation of these systems, as well as the magnetocaloric effect.

A late addition to our research program was the modeling of complex physiological systems. This involves nanoscale fluid flow as well as complex networks of biochemical reactions. We have been working to develop computational models for mesoscale physico-chemistry and models for relating metabolic response to basic physiological data. One of the systems that we are interested in modeling is the organ of corti of the inner ear and other structures that involve hair cells. The inner ear involves complex fluid flow patterns and acoustic streaming in the basic transduction channels. This is very low Reynolds number flow. While demand is high for a 'virtual liver' i.e., a mathematical model for the complex metabolic reactions that could be studied for possible toxicological control, science is far from understanding the complex web of chemical reactions involved. One of our long-term goals is to piece together such a practical system.

Accomplishments/Findings/Deliverables

Films of ceramic ferroelectric materials are important for various uses. The ability of such materials to be polarized in opposite directions means that it can be used in non-volatile ransom access memories. In addition, such films are frequency agile due to the nonlinear field dependence of the dielectric constant. This makes them attractive for a variety of uses, e.g., in microwave applications. AFRL is fabricating a promising ferroelectric film. barium strontium titanate ($BaSrTiO_3$), by pulsed laser deposition. They are particularly interested in materials that exhibit new properties via controlled synthesis and assembly on the nano scale. Process control is crucial for film quality. The nanostructure of the deposited film can be controlled by varying PLD deposition parameters such as: chamber ambient gas, laser beam energy density, substrate temperature, deposition time, target system parameters, pulse repetition rate, pulse length, substrate-to-target distance, laser beam wavelength, target-substrate relative geometric arrangement and post-deposition annealing. Our research has emphasized understanding the complete physical modeling and theory necessary for a reduced-order mathematical model that will allow an eventual automatic control of this process. The main difficulty has been a gap in the understanding of basic science of the plasma plume generated by the laser ablation in the PLD chamber. The plume is very dynamic (supersonic, high-temperature, shocks, etc.) especially in the presence of a reacting background gas. We have developed mathematical models that include most of the basic physics governing the plume, collaborating closely with AFRL experimental scientists who are generating data files as part of the verification process. There are reports from both experiment and simulation that show exagerated forward-direction to the plume. We have an analytic model to explain this. We have been analyzing the time-of-flight signatures of ablation plumes recorded by AFRL scientists. These recordings have many complicated and mysterious features, We have identified several sources for these phenomena, including reflected shocks, shock-induced vorticity, charge separation and chemical species separation. We have also been using simplified material systems (Cu, CuO, CeO, BaO) as tests of the basic model. Details will be found in a series of papers authored by Svobodny, Biggers, and others. Further information can also be found at the website: http://www.math.wright.edu/MS/AppliedMath/research.html . Deliverables to AFRL include: a comprehensive PLD modeling guide, and a suite of computer programs for the use of AFRL scientists to allow immediate analysis of experimental results in pulsed laser ablations. This software includes ABSOL, a computer program that relates laser energy to plume measurements and film measurements, and PLD1DTOF and PLD2DTOF computer programs that explain TOF measurements in terms of blast models and related simple models of plume development.

The composition of the plume depends on thermochemistry. The mechanism of oxide production and resultant possible chemiluminescence remains controversial. Making this problem more difficult is the dearth of any data on the physical chemistry of these reactions. For this reason, we have been doing density functional calculations to determine the energy curves of possible reactions. The next step is to incorporate this model with the fluid mechanics into a master model of the reacting plume. We have analyzed the plume from the point of view of competing scales. For expansion into a higher pressure ambient, there are two competing models for one dimensional plume expansion. The *blast wave model* describes the self-similar radial expansion when energy is deposited at a point at $t = 0$. The *normal shock model* describes one-dimensional motion peaked in a particular direction. Following closely on experiments, special solutions can be constructed each valid on scales widely separated from others. Computations also show this difference. We have shown that both scaling regimes can be incorporated in a single model with an adjustable 'geometric' parameter. In some situations the plume dynamics cannot be described by a one-dimensional model. The plume dynamics in the presence of a temperature gradient is one such example, particularly in situations when the plume is seen to be extremely forwarded-directed. In experiments where substrate heat was isolated as a control, the plume showed an increase in size and emission intensity that was similar to the effect of increasing laser beam energy. However, in a vacuum, the heater produced no discernible change to the plume. Thus, the effect was due to molecular kinetics and not to radiative heat transfer. Moreover, this has been reproduced in our numerical computations, and has been explained via an analytical model. Our latest work seeks to extend this analysis to include the important chemical reactions that play an important role in the plume dynamics. We are continuing to develop our continuum computational model as well as to experiment with using Monte Carlo simulations on the reacting plume.

Nanocluster synthesis is important for many material systems and PLD provides one pathway. Of course, we are interested in understanding how this synthesis depends on the dynamics of the plume. We have been investigating how the use of an ambient pressure control changes the plume dynamics and how this in turn effects the nanocluster formation. In a high-pressure ambient, there are stronger shocks and quicker relaxation to the diffusion stage. This makes it likely that nanoclusters form in the plume before they reach the substrate.

Nano-structured magnetic materials have a complex mesoscopic crystalline structure, similar to new magnetic materials but that have been considered for use in refrigeration. The materials used in these magnets are superparamagnetic nanoclusters: that is, they consist of magnetic nano-sized particles imbedded in a matrix which may or may not itself be magnetic. Most quantitative work on the magnetocaloric effect has not been very accurate. This is because the interactions of the spin thermodynamic system with the underlying elastic vibrations has not been taken into account. The full interaction is very difficult, since, even under the assumption of linear elasticity and linearized magnetism, where the excitations are phonons and magnons, the magneto-elastic interaction is inescapably non-linear. We have looked at simplified models, where the magnon and phonon branches of the spectrum combine to give a new type of excitation that is used for the quantum statistics. What is very interesting is that these *quasi-phonons* can have a spectral branch that dissapears near a phase transition. This could mean that latent heat can be stored in these excitations. These materials are being prepared at Wright State. Besides being designed to match a certain model of magnetoelasticity, this material system is an example of a mesoscopic system with significant interactions in the nanometer-micron range. The manipulation and process control of the spatial and temporal structure of matter over mesoscopic distance scales, especially near phase transitions, has been identified as important to the long-range war-fighting capabilites of the Air Force.

Personnel Supported

Besides the PI, no personnel were supported on this grant. However, the PI has collaborated with colleagues at Wright State and other Institutions, and has been aided by students who are supported through other grants.

Publications

Technical Reports

1. Annual Scientific Report September, 2004
2. Annual Scientific Report September, 2002
3. Annual Scientific Report September, 2003

Scientific Publications

1. Modeling, computation, and control of pulsed laser materials processing, accepted for publication in International Journal of Numerical Analysis and Modeling
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8. Ablation Plume Shock Wave in a Temperature Gradient Paper Number (with R. Biggers) AIAA-2005-0313

Interactions

Conference meetings

March 02 Annual Meeting of American Physical Society, Indianapolis, IN; April 02 SPIE High Power Laser Ablation, Taos, NM; May 02 AFOSR contractor's meeting; October 2002 Fall Meeting of American Physical Society, Ohio Section; January 03 AIAA Aerospaces Meeting, Reno, NV; March 03 Annual Meeting of American Physical Society, Austin, TX; May 03 AFOSR contractor's meeting, Eglin, FL; May 04 AFOSR contractor's meeting, Dayton, OH; January 05 AIAA Aerospaces Meeting

AFRL interactions

Rand Biggers of MLPS-AFRL was the main collaborator of the PI. This collaboration is in the process of producing several research monographs, including a substantial guide to the modeling of PLD. The PI has also interacted with Air Force personnel interested in picosecond and femtosecond pulsed laser interaction with matter, and with scientists interested in fabricating ferroelectric materials. The PI has interacted on several occasions with scientists from AFRL Air Vehicles Directorate and has written research proposals. The PI has also interacted with the Human Effectiveness directorate concerning physiological modeling, systems biology, and virtual metabolism. This collaboration, which supports the AF lab investigating toxicity, and survivability in toxic environments, has been successful in attracting funding.

Other interactions

Hosted collaborators from University of Utah, Iowa State University, Charles University, and New Jersey Institute of Technology. The PI co-hosted in December03 a workshop on Mathematical Modeling and Scientific Computation in Science. The PI has visited School of Computational Science & Information Technology at Florida State University. In talks, we emphasize the wide applicability of our results to areas outside of materials processing.

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